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CONCERNING THE ELECTROSYNTHESIS OF HYDROGEN
PEROXIDE AND PEROXODISULFATES. SECTION II:
OPTIMIZATION OF ELECTROLYSIS CELLS, USING AN
ELECTROLYZER FOR PEROXODISULFURIC ACID AS AN
EXAMPLE

Martin Schleiff, Wolfgang Thiele, Hermann Matschiner

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Concerning the Electrosynthesis of Hydrogen Peroxide and Peroxo-disulfates

by Martin Schleiff, Wolfgang Thiele and Hermann Matschiner

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Part II: Optimization of electrolysis cells, using an electrolyzer for peroxodisulfuric acid as an example.

Communication from the VEB Eilenburger Chemie Werk, concern of the Combine VEB Chemische Werke Buna, management area Research and Development, and of the Martin-Luther University Halle-Wittenberg, departments for Mathematics and Chemistry.

The calculation of electrochemical reactors on the basis of balance equation systems [1] is still limited at present to simple cell geometries. In addition it requires the knowledge of a large number of material- or system-related parameters which, in real cases, are generally unknown and which can only be determined with great experimental expenditures. On the other hand, in the application of combined statistically and physically based mathematical models, results can be obtained in a short time with relatively small experimental expenditures for the determination of the empirical constants introduced into the model, which can make an important contribution toward optimization and perfection of electrolyzers of complicated engineering construction.

Such models were developed in the VEB Eilenburgwer Chemie Werk and have been used for some years successfully for the optimization of engineering and economic characteristic values [2]. We now plan to present here the model of an electrolyzer for peroxodisulfuric acid and to analyze it mathematically. Its application for engineering and economic optimization will be investigated in detail.

* Numbers refer to foreign pagination.

Process-theoretical model

In the modelling of the electrolyzers we shall dispense with a description of the cell design as well as the processes in the interior of the cell; for this we refer to part I [3]. We use only relations between the external process variables to set up the model, whereby the effect of thermal-engineering parameters is based on physical causes. The variables, which can be taken into account to set up the models for a fixed cell design and constant electrolyte composition, are the current strength I , the dosing speed and the external temperatures, i.e., the cooling water temperature ϑ_1 and the ambient temperature ϑ_2 . In the subject application case for the production of hydrogen peroxide it makes sense to vary the dosing rate and the current strength always in the same proportion to each other, so that the final concentration of the peroxodisulfuric acid falls into the range most favorable for the subsequent hydrolysis stage. Thus the dosing rate is not an independent variable; the effect of the current strength then contains the effect of the changing flow velocity and the average dwell time. The relations between the remaining independent variables and the quantities, which describe the electrolysis process, such as electrolysis temperature ϑ , cell voltage U , current yield η and specific electric energy consumption E_s are then given sufficiently accurately by the simplified interrelation model shown in figure 1.

To set up the mathematical model it is now necessary to describe each interrelation by a function, using data obtained by measurements. In order to keep interferences with the operational electrolysis process as small as possible, we only made measurements here which for fixed external temperatures $\vartheta_1^{(0)}$ and $\vartheta_2^{(0)}$ for the different values of current strength I determined the associated values for the electrolyte temperature ϑ , the cell voltage U and the current yield η . In place of the current strength I we shall use as variable in the following the current density i referenced to a reference area F (e.g., the cathode area). One can now represent the desired target quantities in a purely statistical

way as functions of the starting variables. It turns out, however, that it is exactly the stepwise buildup of the model suggested by the interrelations with the use of some physically justified relations, which leads to an essentially more informative model.

Initially the current yield η and the cell voltage U are introduced as functions of the current density i for fixed external temperatures $\vartheta_1^{(0)}$ and $\vartheta_2^{(0)}$

$$\eta = x_1^{(0)} i^2 + x_2^{(0)} i + x_3^{(0)}, \quad U = a_0 i + b_0 \quad (1)$$

whereby the coefficients $x_1^{(0)}, a_0, b_0$ are determined from the measured values by the method of least squares. The cell temperature is determined with neglect of reversible thermal changes from a simplified energy balance

$$\kappa_1(\vartheta - \vartheta_1) + \kappa_2(\vartheta - \vartheta_2) = F i [U - \eta(E_1^0 - E_2^0) - E_2^0] \quad (2)$$

in which κ_1 and κ_2 denote the heat transfer coefficients, referenced to one electrolysis cell, in the cooling fluid or in the environment /680 and $E_1^0 = 2,2 \text{ V}$, $E_2^0 = 1,2 \text{ V}$ the standard EMK of the anodic peroxide or oxygen formation. To make possible a simple solution for ϑ , we consider η here as approximately constant with a value of 0.7 and use for U the relation from (1), whereby the temperature dependence of U is thus neglected. Under these assumptions it is possible to determine the values $\kappa_1^{(0)}$ and $\kappa_2^{(0)}$ for the electrolysis cell considered with the method of least squares from the values measured for ϑ .

The solution of (2) for ϑ produces

$$\vartheta = \frac{F i (a_0 i + b_0 - 1,9)}{\kappa_1 + \kappa_2} + \vartheta_3 \text{ mit } \vartheta_3 = \frac{\kappa_1 \vartheta_1 + \kappa_2 \vartheta_2}{\kappa_1 + \kappa_2} \quad (3)$$

whereby the designation ϑ_3 is used as abbreviation for the average external temperature. In order to now obtain the current yield and the cell voltage U as functions of the two variables i and ϑ , the expressions

$$\eta = f_1(i) + f_2(\vartheta), \quad U = g_1(i) + g_2(\vartheta) \quad (4)$$

are set up with separated variables. The summands depending on ϑ

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are determined by integration from the approximately known temperature coefficients

$$\partial\eta/\partial\vartheta = -0,0006 \vartheta, \quad \partial U/\partial\vartheta = -0,01 \quad (5)$$

The summands depending on i can be determined in that one demands the agreement of (4) and (1), if one inserts in (4) the external temperatures $\vartheta_1^{(0)}, \vartheta_2^{(0)}$ and the heat transfer coefficients $\kappa_1^{(0)}$ and $\kappa_2^{(0)}$ used as basis in formulae (1). If in the thus obtained functions for η and U the temperature ϑ is substituted as per (3), one finally obtains η and U as functions of the starting variables in the form

$$\begin{aligned} \eta &= x_3 + x_2 i + x_1 i^2 + x_4 i^3 + x_5 i^4, \\ U &= c i^2 + a i + b \end{aligned} \quad (6)$$

with the coefficients

$$\begin{aligned} x_1 &= x_1^{(0)} + \alpha B^2 (1 - D^2) + 2 \alpha A (\vartheta_3^{(0)} - D \vartheta_3), \\ x_2 &= x_2^{(0)} + 2 \alpha B (\vartheta_3^{(0)} - D \vartheta_3), \\ x_3 &= x_3^{(0)} + \alpha (\vartheta_3^{(0)2} - \vartheta_3^2), \\ x_4 &= 2 \alpha AB (1 - D^2), \quad x_5 = \alpha A^2 (1 - D^2), \quad \alpha = 0,0003 \end{aligned} \quad (7)$$

and

$$\begin{aligned} c &= \beta A(1 - D), \quad a = a_0 + \beta B(1 - D), \\ b &= b_0 + \beta (\vartheta_3^{(0)} - \vartheta_3), \quad \beta = 0,01 \end{aligned} \quad (8)$$

with the abbreviations

$$A = \frac{F a_0}{\kappa_1^{(0)} + \kappa_2^{(0)}}, \quad B = \frac{F(b_0 - 1,9)}{\kappa_1^{(0)} + \kappa_2^{(0)}}, \quad D = \frac{\kappa_1^{(0)} + \kappa_2^{(0)}}{\kappa_1 + \kappa_2}$$

If η and U are known, one obtains the specific electric-energy consumption as

$$E_s = c_0 U/\eta \text{ mit } c_0 = 2 F_0/M = 276,26 \text{ A} \cdot \text{h/kg} \quad (9)$$

For better visualization of the formulae obtained we have shown in figure 2 the current yield and the specific electric-energy consumption as a function of the current density for different values of the cooling-water temperature ϑ_1 for $\vartheta_2 = 20^\circ \text{C}$. In addition we have plotted the current yield-current density functions for constant electrolyte temperatures ϑ . The plotted curves are based on values which are encountered in a commercial electrolyzer for peroxodisulfuric acid and namely

$$\begin{aligned} \theta_1^{(0)} &= 5^\circ \text{C}, \quad \theta_2^{(0)} = 20^\circ \text{C}, \quad x_1^{(0)} = -0,45 \cdot 10^{-6} \text{A}^2 \cdot \text{m}^4, \\ b_0 &= 3 \text{V}, \\ x_2^{(0)} &= 10^{-3} \text{m}^2/\text{A}, \quad x_3^{(0)} = 0,2, \quad a_0 = 0,7 \cdot 10^{-3} \text{V} \cdot \text{m}^2/\text{A}, \\ F &= 0,8 \text{m}^2, \quad z_1^{(0)} = 100 \text{W/K}, \quad z_2^{(0)} = 25 \text{W/K} \quad (10) \end{aligned}$$

Engineering-economic optimization model

The optimum operating point of a facility is determined not only by the engineering-physical data, but also to a considerable extent by the economic data. In order to set up the associated mathematical model, an analysis of all the expense types affecting all the total costs of an electrolysis process is necessary; these must be prepared in the form of cost functions.

Among the fixed costs, which are independent of the current density and the amounts produced we count the basic equipment expenditures for the electrolyzers and secondary facilities in the form of annual amortizations and the fixed operating costs, such as for example wages and salaries. The variable costs include:

- the costs for the electrolysis current; they are proportional to the power consumption and are calculated with an electric-power cost d_1 and the efficiency d_2 of the DC current generation as

$$K_1 = c_1 d_1 U i / d_2 \quad \text{with } c_1 = 8600 \text{ h/a}$$

- the costs for general repairs whose frequency increases proportionally to the electrolysis current

$$K_2 = d_3 i$$

- the energy costs for the cooling facility which are proportional to the quantity of heat which is picked up by the cooling medium along a square meter reference area F

$$K_3 = c_1 d_1 d_4 z_1 (\theta - \theta_1) / F$$

(here d_4 is the ratio of the expended electric power to the quantity of heat picked up and removed by the cooling fluid).

- the costs proportional to the annual production quantity m /681

$$K_4 = d_5 m, \quad m = c_2 i \eta, \quad \text{with} \quad c_2 = c_1/c_0 = 31,13 \text{ kg/aA}$$

Thus the total costs encountered per year per square meter of electrolyzer area are

$$K = K_0 + c_1 d_1 U i / d_2 + d_3 i + c_1 d_1 d_4 z_1 (\theta - \theta_1) / F + c_2 d_5 i \eta \quad (11)$$

The analysis of the total costs for a realistic engineering electrolyzer showed that the costs K_1 for the electric power with almost 60% represent the greatest share, followed by the production-proportional costs with almost 25%. The target functions important for an optimization of the electrolysis process now are the specific costs k per unit quantity produced

$$k = K/m = K/(c_2 i \eta) \quad (12)$$

and the operating result per year and per square meter of electrode area

$$BE = d_6 m = K \quad (13)$$

whereby d_6 denotes the calculated price for the electrolysis product.

For better visualization all target functions are shown in figure 3 as functions of the current density. Here those values were used as bases for the economic parameters, which can be encountered in a production persulfuric acid electrolyzer, and namely

$$\begin{aligned} K_0 &= 1000 \text{ M/a} \cdot \text{m}^2, & d_1 &= 0,15 \text{ M/kW} \cdot \text{h}, \\ d_3 &= 0,3 \text{ M/a} \cdot \text{A} \\ d_2 &= 0,9, & d_4 &= 0,4, & d_5 &= 0,1 \text{ M/kg}, & d_6 &= 0,6 \text{ M/kg} \end{aligned}$$

One can see in the figure that the various target functions always have an optimal value which, however, is assumed for different current density values.

Evaluation of the model

It was shown that the derived model reflects the electrolysis process in the range, interesting from a production standpoint, between 900 and 1500 A/m² with sufficient accuracy. It should not be extrapolated beyond this range since for lower current densities, e.g., the linear expression (1) for the cell voltage is no longer correct and since for higher current densities, e.g., the expression (6) for the current yield at constant electrolyte temperature decreases again while from a physical-chemical standpoint one could expect that it approaches a limiting value.

Between the optimum current density values for the here considered target functions of the process-theoretical and production-economic optimization, there exist a few general relations which are always valid independently of the individual parameter values. They either satisfy the inequality chain

$$(i_{opt})_{E_s} < (i_{opt})_{\eta} \leq (i_{opt})_k < (i_{opt})_{BE} < (i_{opt})_m \quad (14)$$

or the inequalities

$$\begin{aligned} (i_{opt})_{E_s} < (i_{opt})_{\eta} < (i_{opt})_m, \quad (i_{opt})_k \leq (i_{opt})_{\eta}, \\ (i_{opt})_k < (i_{opt})_{BE} < (i_{opt})_m \end{aligned} \quad (15)$$

We shall dispense here with proofs for all inequalities; we shall only examine in detail the in practice particularly interesting inequality $(i_{opt})_k < (i_{opt})_{BE}$. From the necessary extreme-value requirement for $k = K:m$ there follows $K'm - K m' = 0$. If one solves this for m' and inserts this into the derivative of BE, one obtains

$$\begin{aligned} BE' &= d_6 m' - K' = d_6 \frac{K'm}{K} - K' = \frac{K'}{K} (d_6 m - K) = \\ &= \frac{K'}{K} BE > 0 \end{aligned} \quad (16)$$

If we assume that the operating result BE is positive, which only seems to make sense here, then BE' is positive at $(i_{opt})_k$ i.e., the operating result is a function still growing there.

Thus if in an arbitrary electrolysis facility one is operating at a minimum cost setting, then the operating result can always be still increased. Reversely the setting for maximum yield is not the most cost-effective one. Therefore a new facility should be laid out such that the projected production quantity is reached at a cost minimum, but that a variation of the current density is possible in the range between the extreme values for both target quantities.

The optimum current density values, whose general setting with respect to one another has just been characterized, can be easily calculated by an approximation method in every real case via the necessary extreme value conditions. Figure 4 shows for the parameter values, used as example here, the optimum current densities and the relative function values at the optimum, referenced to $\vartheta_3 = 8^\circ\text{C}$, as a function of ϑ_3 or ϑ_1 .

After the optimum for a given parameter value has become known, it is often interesting to find out how the optimum changes when the parameter value is changed. Thus one must calculate the change of the implicitly determined optimum current density as the result of the necessary extreme value requirement and of the extreme value of the function. Since only small changes of the parameter are considered, so as not to go beyond their validity range of the developed model, the development of a power series provides a useful approximation solution for this purpose. Thus we calculated in this way for the concrete case of a few parameter values the relative changes of the optimum current density listed in table 1 or the optimum target function value for a 1% change of the corresponding value of the parameter. One can see that a change of the starting parameters affects the operational result considerably more than the specific costs. Furthermore it becomes clear that the location of the optimum current density values depends greatly on the economic characteristic values, such as the price of electric power and the price of the product. It can also be seen that a change of the statistically determined coefficients $x_1^{(0)}, x_2^{(0)}, x_3^{(0)}$ of the current yield-current density function substantially affects the location and

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the magnitude of the optimal values. Thus the dependence of these coefficients on design quantities is to be investigated in the continuation of the present project.

Now special interest must still be given to the dependence of the target functions on the physical-engineering parameters found in the model. The effect of the cooling-water temperature θ_1 on some target functions, as can be seen from figure 4, shows that a low cooling-water temperature is the most favorable even if the cooling costs are taken into account. Since limits are set for a lowering of the cooling-water temperature by the performance of the cooling facility, other measures for lowering the electrolyte temperature are important. Therefore we shall next consider the effect of the heat transfer coefficient α_1 . An improvement of the heat transfer properties increases the costs for the construction of the electrolysis cell; as an example let us assume that the fixed costs can be determined by the formula

$$K_0 = K_{00} + d_0 (\alpha_1 - \alpha_1^{(0)}) .$$

For $d_0 = 1, 2, 3 \text{ M} \cdot \text{K/a} \cdot \text{m}^2 \cdot \text{W}$ we calculated with this assumption the minimum specific costs and the cost-optimal current density as shown in figure 5. One can see that the minimum costs first decrease with increasing heat transfer coefficient α_1 , but then increase again. The thus determined best value for the heat transfer coefficient provides information for an optimum design electrolysis cell from a thermal-engineering standpoint.

One can obtain an improvement of the heat transfer properties especially easily by an enlargement of the cathode surface which essentially coincides with that of the cooling area. If one introduces an enlargement factor p into the model and if one takes into account the dependence of the fixed costs of p in an expression corresponding to formula (17), then the calculation of the specific costs as a function of p provides results which are similar to those shown in figure 5. From this one obtains information about an optimally designed electrolysis cell with

respect to the cathode area.

The results and conclusions derived from the model show that even with a relatively simple model and with relatively low measurement expenditures one can derive important information not only for optimization of the operating conditions of an existing electrolyzer, but also for continued design developments and that here an analysis of the formally mathematical structure can already provide information about which changes of the target functions are at all compatible with the design of the model. The here presented model makes possible with continuing scientific understanding of the electrolysis process a stepwise continued development in that further interrelation expressions are explained physically and that the effect of engineering design parameters is revealed by the model. In this way the extrapolation capability can be improved and an optimization of the electrolyzer with respect to additional parameters becomes possible.

Summary

A combined statistically and physically justified model is presented and evaluated with an example of an electrolyzer for peroxodisulfuric acid. An engineering economic optimization is made with the aid of an analysis of all costs affecting the electrolysis process. By means of a mathematical analysis of the model we drew generally applicable conclusions concerning the change in position of the optimum with respect to the various investigated target functions as the result of changes of the individual design-caused and economic parameters.

symbols	explanation
a_0, b_0	coefficients of the cell voltage function (starting cell)
a, b, c	coefficients of the cell voltage function (arbitrary cell)
BE	operating result per cell and per year
cj	conversion factors (constants)
dj	cost factors
E_s	specific electric power consumption
E_j^0	standard EMK
F	reference area of one cell
F_0	Faraday constant
I	current strength
j	current density (referenced to the cell area F)
K	total costs per cell and year
K_j	cost shares per cell and year
k	specific costs
M	molar mass
m	production quantity per cell and year
U	cell voltage
$x_j(0)$	coefficients of the current yield functions of the starting cell
r_j	coefficients of the current yield functions of an arbitrary cell
ϑ	electrolyte temperature
$\vartheta_j^{(0)}$	external temperatures (starting cell)
ϑ_j	external temperatures (arbitrary cell)
η	current yield
$x_j(0)$	heat transfer coefficients (starting cell)
x_j	heat transfer coefficients (arbitrary cell)

References

- [1] Hertwig, and Thiele, R.: Chem. Techn. 29 (1977) 76.
- [2] Thiele, W.: thesis , Martin-Luther University Halle-Wittenberg 1977
- [3] Thiele, W. and Matschiner, H.: Chem. Techn. 29 (1977) 148.

Submitted on April 6, 1977.

Addendum

In part I of this series, which was published in Chem. Techn. 29(1977) 148 to 154, the part of the table belonging to figure 1 was accidentally not printed. The corresponding values are listed in the following. The table must be added directly below the figure 1 and constitutes a unit together with it.

current- A/electrolyzer	300	1000	7000	1000	12000	14000 ~
capacity A/diaphragm	≈ 75	100	25	500	500	550
in A/m ² area	≈ 2000	4000	5000	4000	7000	8000
requirement						
current yield in %	≤ 60	70	74	72	70	78
ref. to comparable						
H ₂ S ₂ O ₈ conc.						
of 27.....33%						
cell design	orig. W cell	W cell as "fast electr." per Baum/Teicher	D-W cell as per Müller	USSR electr.	E electr. EZ I	E electr. EZ II
original patent		DT567542	DT975825	SUL67492	DL27961	DL99548
init. appl. date	1905	1930	1951	1957	1963	1972

W = Weissensteiner cell D = Degussa cell E = Eilenburger cell

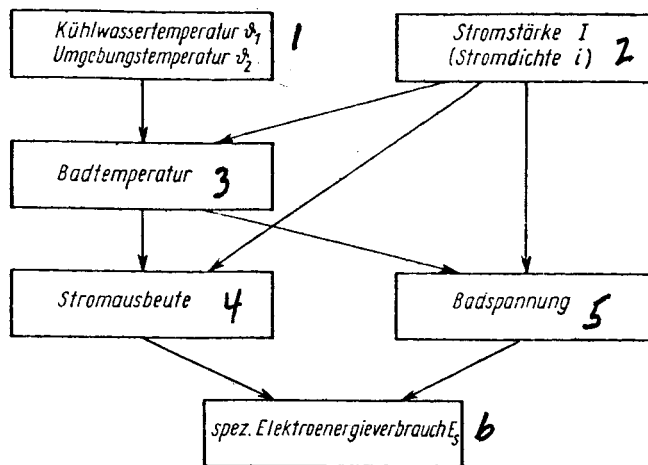


Figure 1 Interrelation model

- 1- cooling water temperature θ_1 ambient temperature θ_2
 2- current strength I (current density i)
 3- bath temperature 4-current yield 5- bath voltage
 6- spec. electric power consumption E_s

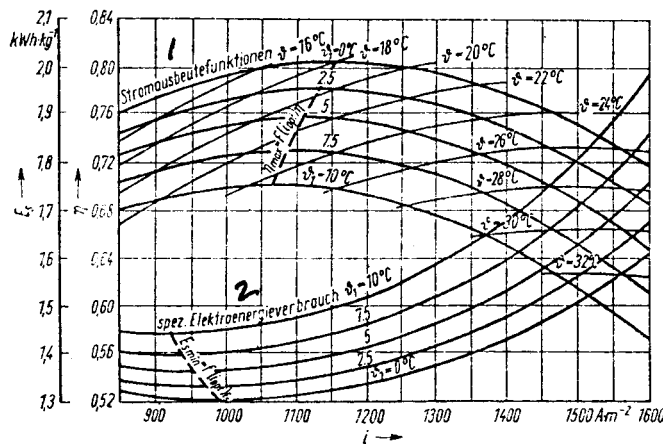
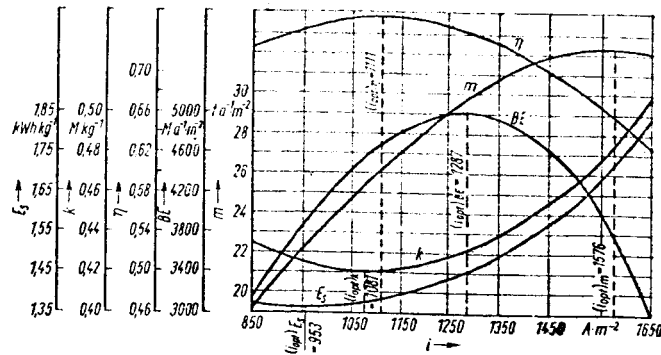


Figure 2 Relation between current yield and specific electric power consumption and the current density for different cooling water temperatures θ_1 ($\theta_2(0) = 20^\circ\text{C}$)

- 1- current yield functions
 2- spec. electric power consumption



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Figure 3 Current density dependence of the investigated target functions ($\vartheta_1^{(0)} = 5^\circ\text{C}$, $\vartheta_2^{(0)} = 20^\circ\text{C}$)

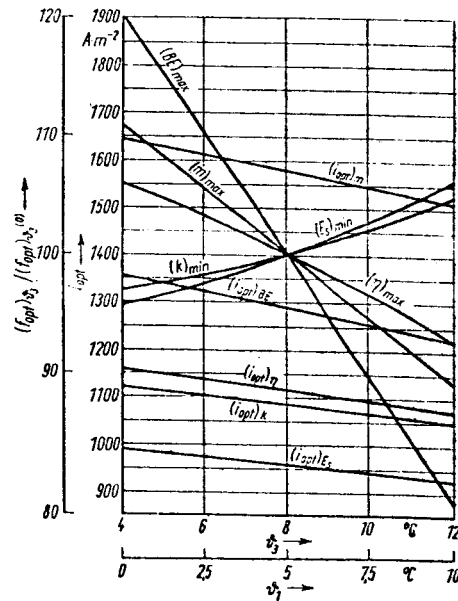
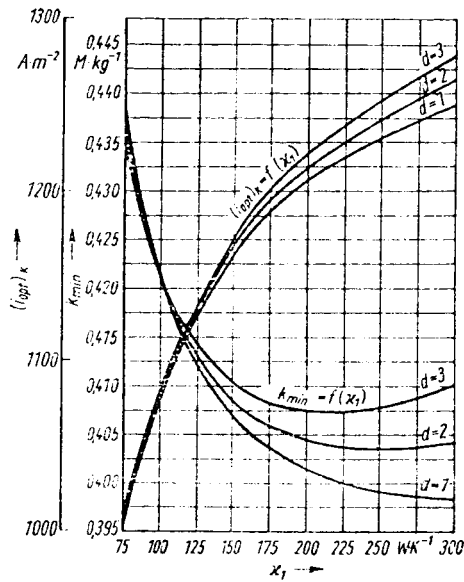


Figure 4 Dependence of the relative extreme function values, referenced to the starting temperature $\vartheta_3^{(0)}$ and of the optimal current densities on the external temperatures ϑ_3 and the cooling-water temperature ϑ_1 (for $\vartheta_2 = 20^\circ\text{C}$)



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Figure 5 Dependence of the minimal specific costs and the cost-optimal current densities on the heat transfer coefficient for cells with design changes having different fixed cost factors $d_0 = 1, 2, 3$ ($\theta_1 = 5^\circ\text{C}$, $\theta_2 = 20^\circ\text{C}$)

	$x_1(0)$	$x_2(0)$	$x_3(0)$	d_1	d_5	d_6	K_0
$(i_{opt})_{BE}$	-1.059	1.219	0.095	-0.246	0.051	0.306	—
$(i_{opt})_k$	-0.811	0.817	-0.006	-0.076	—	—	0.075
$(BE)_{max}$	-2.973	5.135	0.798	-1.684	-0.592	3.552	-0.199
$-k_{min}$	-0.531	1.092	0.202	-0.639	-0.237	—	-0.093

Table 1 Relative change of the optimal current density and the optimal target function values during changes of the parameter value by 1%.

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16. Abstract The model is presented of an electrolyzer for peroxodisulfuric acid, and it is analyzed mathematically. Its application for engineering and economic optimization is investigated in detail. The mathematical analysis leads to conclusions concerning the change in position of the optimum with respect to the various target functions due to changes of the individual design-caused and economic parameters.			
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